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PHASE SEPARATION UNDER A CONCENTRATION GRADIENT

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Binary mixtures have been investigated for a long time to understand the dynamics of phase separation. One of the important results of these studies is that the process of phase separation can be identified as a slow nucleation (N) or a fast spinodal decomposition (SD) /1/ depending on the volume fraction ϕ of the two components of the binary mixture. For systems with small ϕ , there is a clear distinction between a majority and a minority phase. Phase separations in such a system proceed as the nucleation of the minority phase from the majority phase. The phase separating domains are therefore isolated droplets. However, when ϕ is close to 0.5, there is no clear distinction between a majority and a minority phase. In this case, phase separations proceed by SD which is characterized by the formation of an inter-connected structure.

For a spatially homogeneous system, ϕ is a constant. Only one of the above two processes can take place. However, if there is a spatial variation of composition (concentration gradient) in the binary mixture where $\phi(z)$ is a function of position z , both nucleation and spinodal decomposition can occur simultaneously at different location of the sample. One will expect to find SD at places where ϕ is close to 0.5. For such an inhomogeneous system, it would be interesting to know if new processes, other than N and SD, will occur during the phase separation process.

Intuitively, the concentration gradient λ ($= dc(z)/dz$ or $d\phi(z)/dz$) of the inhomogeneous system plays an important role in determining the phase separation process. For a binary mixture undergoing phase separation near the critical point, a natural unit for concentration gradient will be $\lambda_0 = \Delta c/\xi$ where Δc is the difference in concentration of the two final phases and ξ is the correlation length of the system. Since the concentration gradient which can be supported by the equilibrium system is the transition interface /2/ between the two phases, one can distinguish between the case of a weak concentration gradient when $\lambda \ll \lambda_0$ and the case of a strong concentration gradient when $\lambda \sim \lambda_0$.

One of the early studies of phase separations under a weak concentration gradient was carried out numerically by

Kolb et al. /3/ to understand the fractal structure observed in experiments of silver deposits in polyimide films. In this lattice simulation study, they found that the concentration profile is conserved during the phase separation as long as the domains are much smaller than the gradient length scale and no specific effects of the weak concentration gradient is observed. Recently, in a phase separation experiment of a binary mixture under a weak linear concentration gradient, Beysens et al /4/ found that nucleation and spinodal decomposition can be observed simultaneously at different location of the sample. Figure 1 is an illustration of the occurrence of both N and SD and its relation with the concentration gradient.

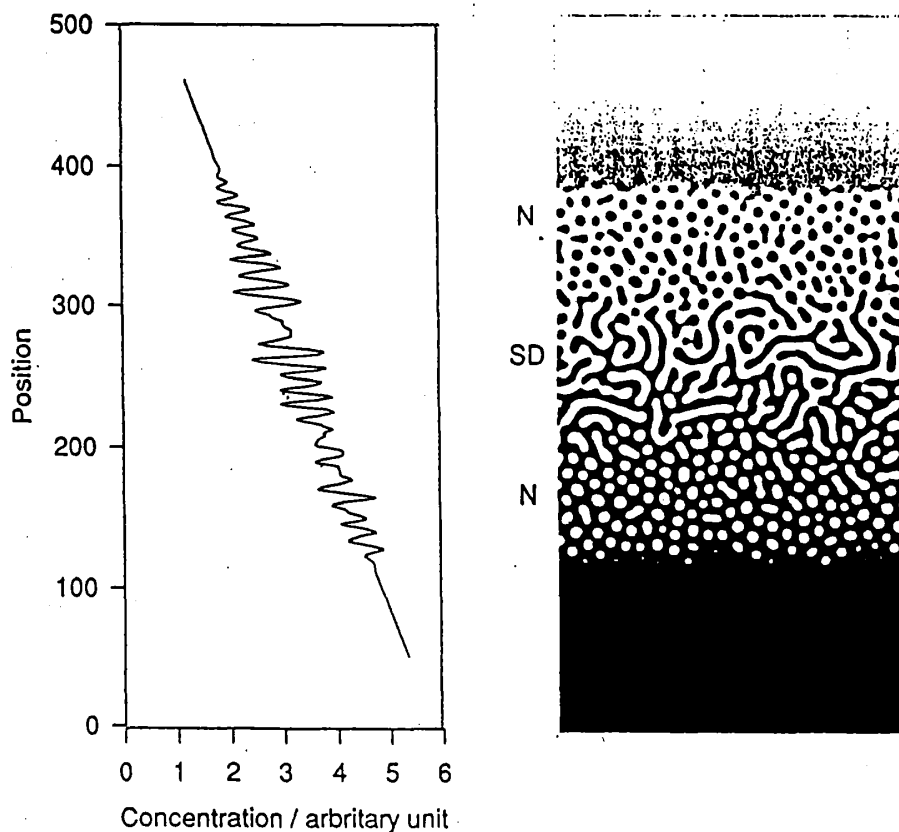


Fig.1 Phase separation under a concentration gradient. Simulation results of a system similar to that of Ref. 5.

Similar to Kolb et al's findings, the growth law of the N and SD observed in Beysens et al's experiments do not seem to be affected by the presence of the weak concentration gradient. The growth of the phase separating domains are still characterized by the $t^{1/3}$ and t^1 time dependence for N and SD respectively. However, the concentration gradient is not conserved during the phase separating process. This non-conservation of concentration gradient is due to the large

scale structure formation of the phase separating domain which occurs at late time. A recent computer simulation for late time growth /5/ of phase separation under a concentration gradient also confirmed this non-conservation.

The case of a strong concentration gradient was recently studied by Chan /6/ experimentally. The transition interface of a phase separated binary mixture was used to produce the strong concentration gradient with $\lambda \sim \lambda_0$. Obviously, no phase separation will take place if $\lambda = \lambda_0$. But for $\lambda < \lambda_0$, the equilibrium transition interface is found to form very quickly through an anisotropic SD during phase separation. The anisotropy is such that the growth of the phase separation domain is slower in the direction of the concentration gradient. One of the most interesting feature of this anisotropic growth is that the small angle light scattering pattern is in the form of an ellipse only during early time. At late times, when this spinodal ring collapses into the forward scattering direction, a diffraction-like scattering pattern is formed as shown in Fig 1. Note that scattering in the form of an collapsing ellipse can also be observed in the phase separation of a binary mixture under a constant velocity gradient /7/.

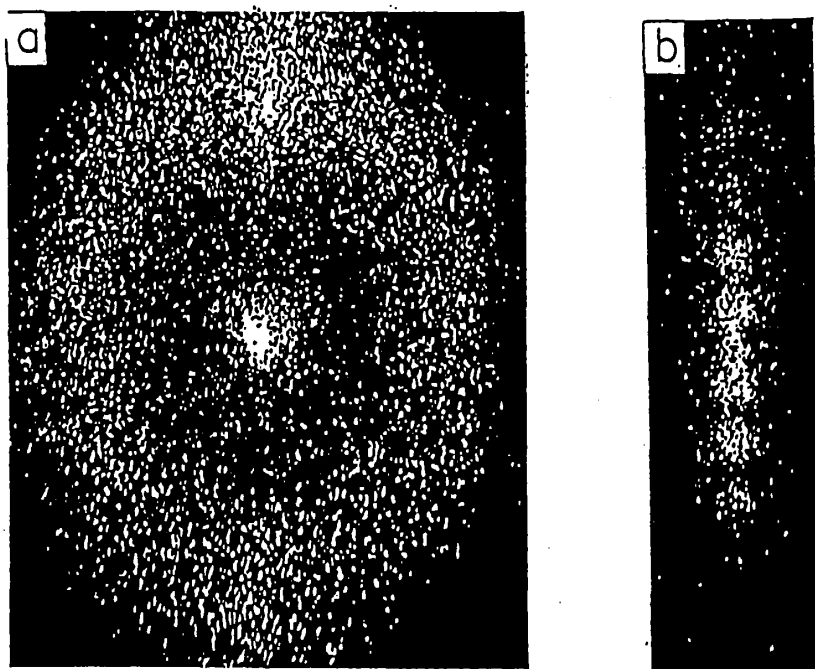


Fig.2 Small angle scattering pattern of phase separation in a strong concentration gradient. a) early time b) late time. For details see Ref.6.

The above description is only a very brief introduction of the recent developments in the understand of the phase separation process of a binary mixture subject to a concentration gradient. Some of the observations in the experiments are still not understood yet; especially in the case of a strong concentration gradient. Flows induced by phase separation are inevitable in most situations /7/. Hydrodynamics might be a very important factor in the understanding of the phase separation experiments.

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